**Atmospheric
Chemistry
and Physics
Discussions**

Interactive comment on "The continental source of glyoxal estimated by the synergistic use of spaceborne measurements and inverse modelling" *by* T. Stavrakou et al.

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4D-Var data assimilation of HCHO and CHOCHO columns is used to estimate missing continental sources of glyoxal in the IMAGESv2 model. Inverse model solutions are primarily validated through comparison of the optimized model estimates to surface observations of glyoxal. The robustness of the solution is tested by additional sensitivity runs with different model setups. It is concluded that the observations and the model are most consistent with an additional secondary source of glyoxal of more than 50 Tg/y. This paper is well written, the subject is timely and the approach is novel. I have questions about a few aspects of the inverse modeling that feel a bit glossed

over and some suggestions for additional analysis to strengthen the presentation of the conclusions. My main suggestion for addition research is to perform a few more inverse modeling tests that begin with higher than a 108 Tg/y source of glyoxal in order to determine if this inverse modeling framework really can provide a bounded estimate from both sides, and to further discuss the implications on tropospheric chemistry of the inferred secondary glyoxal source.

0.0.1 Comments

1. Overall, I would appreciate seeing more in this paper about the inverse modeling approach. Having established a track record of 4D-Var data assimilation with the IMAGES adjoint, I agree there is no need for the authors to repeat content about adjoint model derivation, optimization, etc. Yet, I think they could still provide additional discussion in the introduction that connects this piece to the young, yet growing, body of literature on inverse modeling emissions of chemically reactive species using adjoints of CTMs. Many readers are still skeptical if not dismissive of top-down emissions constraints, so I think further discussion along these lines is warranted.

2. Related to the previous comment, it struck me right away that this is the first adjoint model to consider SOA formation. Nobody has yet applied 4D-Var to SOA, so that could be mentioned in the conclusions and/or abstract.

3. 13601.4: Since ISORROPIA is used in the forward model, has the adjoint of ISORROPIA been made for the IMAGES adjoint model? If so, then that is something specifically worth highlighting, as it would be a very valuable development. If not, please mention which chemical/physical interactions are only included in the forward model.

4. 13595.13: I couldn't help but wonder why the study is limited to continental

sources. Later, there is mention that including ocean sources would be beyond the current scope of the paper, but since adjoint methods can address any number of emissions simultaneously, I just was not sure the reasons for excluding this potentially very interesting piece of the current glyoxal puzzle. It might be better if the reasons are clearly stated up front in the introduction.

5. 13596.4: Why is the missing source necessarily biogenic in origin?

6. 13596.16: "...organic aerosols is another potentially significant source of glyoxal." Yet, this and other papers focus more on SOA as being a sink for glyoxal, especially when considering, as is done here, irreversible SOA formation. So why is SOA being mentioned here as a source?

7. 13597.7: "monthly mean ECMWF reanalysed wind fields" Why use the monthly mean winds if the model takes 24h time steps?

8. Are the NO_x emissions inventories mentioned? NO_x levels will affect lifetimes and inferred emissions. While understandably outside the scope of this article, it could be suggested that NO_x observations potentially provide additional constraints for future inverse modeling studies.

9. section 2.6, which describes the surface glyoxal measurements, is missing. Perhaps it would answer why diurnal variations are not considered for model comparisons to the surface observations.

10. 13605.16: "The thresholds has been adjusted so as to avoid the existence of large disparities in the number of unknowns to be optimized from the different categories." It's not obvious why such disparity would be a problem. Could this be explained further?

11. Results: One of the most important parts of the results section is the attempt to verify or discredit the different inverse modeling solutions. In this manuscript, the

approach is to compare estimates from the various optimized models of surface glyoxal concentrations to observations. The analysis would be strengthened by including quantitative metrics of this comparison for each of the sensitivity analysis. For example, the results of sensitivity studies C-F can be compared to the surface measurements, and the resulting correlations and biases summarized in Table 3, or even as labels in Fig. 8. This would be more satisfactory than the textual summary given in lines 23–25 of page 13608.

Comparison to the HCHO measurements also provides an additional constraint, but since these observations were used in the inverse modeling procedure themselves, the utility of such comparisons for validating the inverse modeling solution is different. Still, it would be nice if analysis similar to Fig. 3 was included for runs C-F. Providing a quantitative metric along with these plots would help (for example the % of the converged cost function owing to HCHO observations), because at first glance it does appear that B is better than A, which would be nice to know.

12. For nonlinear optimization problems, there is no guarantee that the inverse model has converged to a global minimum. The authors could and should explore this possibility by trying an additional inversion or two wherein the initial guesses for the CHOCHO or UVOC sources are much higher than those from Inv A or B. This would demonstrate that the inverse model truly is capable of bounding the value of the missing source. Otherwise, the results could simply indicate a lower bound for the missing sources since the model is constrained to the a priori source levels.

13. Lastly, I think there is room for additional discussion of the impacts of the inferred glyoxal source beyond those on concentrations of glyoxal, formaldehyde and SOA. For example, did the concentrations of species such as NO_x and O_3 change much in the IMAGES model after the inversion? Are these changes con-

sistent with current understandings of tropospheric chemistry?

0.0.2 Minor comments and clarifications

- 13595.27: "lanfd" → "land"

- 13599.11 "by 16%" → "which is 16%"

- 13606.16: "thresholds has been" → "thresholds have been"

- ratio plots: strongly suggest using a traditional blue-white-red plot for these types of values. Otherwise, it can not be quickly determined which changes are increases vs decreases. A log scale may also be more appropriate.

- 13606.25: "biogenic isoprene fluxes" → "biogenic fluxes" because the biogenic emissions sector includes other VOC species, correct?

- 13606.26: "in excellent agrement". Typo in agreement. Also, I would strongly suggest providing the quantitative support for this claim, which would make it all the more impressive.

- 13609.15: "source, major" → "source, a major"

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